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56 Abstract

57 Carbon capture and utilisation (CCU) is an emerging technology with commercial potential to 58 convert atmospheric carbon dioxide (CO₂) into net zero or negative emission products. In microalgae-59 based CCU, microalgae utilize CO₂ and sunlight to generate biomass for commercial applications. This 60 paper reviews the current state of microalgal culture development for CCU and highlights its potential 61 contribution to addressing climate change challenges. Current microalgal culture systems have not been 62 designed for high throughput biomass growth and carbon capture. Raceways, high-rate algal ponds, and photobioreactors are the most widely used for microalgal cultivation at a large-scale. The limitations of 63 64 these systems are related to microalgal growth requirements. Ponds are operated at narrow depth to ensure sufficient light distribution and thus need a large land surface. CO₂ gas needs to be in a dissolved 65 66 form for efficient utilisation by microalgae. Innovative system designs to achieve optimised distribution of light, nutrient, and CO2 utilisation for enhanced biomass production are crucial to achieve large-scale 67 CO₂ capture by microalgae. Data corroborated in this review highlights several innovative techniques 68 to deliver CO₂ effectively and enhance light illumination to microalgal cells. Submerged and internal 69 70 illuminations can enhance light distribution without compromising culture volume and land 71 requirements. CO₂ delivery technique selections mainly depend on CO₂ sources. The carbonation 72 column appears to be the best option regarding efficiency, easy operation, and simple design. The downstream processes of microalgal culture (i.e. harvesting, biomass utilisation, and water reuse) are 73 74 important to make microalgae-based CCU a significant contribution to global carbon mitigation 75 solutions.

76

6 **Keywords**: Microalgae; Carbon capture and utilisation; Carbon dioxide delivery; Light

distribution; Microalgal harvesting; Biomass utilisation.

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77

79 1. Introduction

Global warming is the most urgent existential challenge of our time. Although emission reduction
is critically important, it alone is not sufficient to limit global warming below 1.5 °C (IPCC 2021;
Masson-Delmotte et al. 2018). Governments, corporations, and entrepreneurs worldwide have all joined
the global race to develop direct air carbon capture technologies and carbon sequestration systems
(Boot-Handford et al. 2014; Srinivasan et al. 2021).

85 The last two decades have seen unprecedented research investment to develop carbon capture and sequestration (CCS) technologies. They involve the adsorption of CO_2 from flue gas by solvents 86 87 (e.g. alkyl amines) or solid materials (e.g. CaO) followed by thermal desorption or calcination to obtain concentrated CO₂ for long-term storage (Boot-Handford et al. 2014). Concentrated CO₂ is injected in 88 stable geological features (e.g. depleted oil and gas reservoir) or deep oceans where CO₂ is trapped for 89 multiple hundreds or thousands of years. However, underground or deep ocean CO₂ storage has physical 90 91 and geological complexities, which require a complicated risk assessment and extensive monitoring 92 (Boot-Handford et al. 2014; IPCC 2018). The requirements of transportation, pressurization, and 93 ongoing monitoring also increase the cost of CCS (IPCC 2018; Realmonte et al. 2019). There have been 94 19 large-scale carbon capture and storage projects with the capacity of 40 million tons/year (Kamkeng 95 et al. 2021), which is equivalent to only 0.1 % of the annual CO₂ emissions. Completed in 2021, the 96 world's largest direct air capture plant in Iceland can only collect 4,000 tonnes of CO₂ per year.

Utilisation of the captured CO₂ would eliminate the legacy of carbon storage. Carbon capture and
utilisation (CCU) will shift CO₂ for a cost or a waste product to an opportunity (Srinivasan et al. 2021).
CCU is defined as the conversion of captured CO₂ from point sources or the atmosphere into valuable
lower or zero-emission products such as fuels, chemicals, carbon fibres, biomass, and building materials
(Chandra et al. 2019; Dębowski et al. 2020; Srinivasan et al. 2021). CCU has the potential to contribute
to net-zero and negative emissions depending on the downstream utilisation process (Srinivasan et al. 2021).

104 Microalgae-based CCU is a biological process in which CO₂ is converted to biomass by 105 photosynthesis. The produced biomass can be used to replace non-renewable fossil resources in the 106 production of chemicals, fuels, bioplastics, and agriculture feed. Microalgae are photosynthetic, fastgrowing organisms with short generation times. Some microalgae doubling times can range between 4 107 to 8 hours under optimal conditions. They are capable of fixing CO_2 400 times more efficiently than 108 109 terrestrial plants (Sutherland et al. 2021). Microalgae are versatile organisms that can be cultivated on 110 non-arable land, year-round, and in brackish water, seawater or wastewaters. Microalgae cultivation 111 can contribute to reducing global warming emissions by recycling CO₂ into biofuels or generating 112 value-added products from flue gases. Microalgal biomass is a versatile feedstock that can replace fossil 113 materials for raw chemicals, fuels, and industrial products.

114 Currently, commercial microalgal biomass is mainly used to produce cosmetic and nutraceutical 115 ingredients. Microalgal biomass production is estimated at about 30,000 t/yr of carbon dioxide capture 116 (Morales et al. 2017). This is a relatively small amount when compares to the level of CO₂ that needs 117 to be removed from the atmosphere. The extremely low contribution of a microalgae-based process to 118 carbon capture emphasizes the need to increase productivity and scalability to produce a significant 119 amount of microalgal biomass.

120 Recently reported biomass productivities per unit of land size are limited by the sub-optimal conditions used in the microalgal culture system (Khan et al. 2018). Water, nutrients, light, and CO₂ are 121 fundamental requirements for photosynthesis and microalgal growth. In the current state of the art large-122 123 scale reactors, individual microalgal cells do not have the optimal combination of illuminations (light), nutrients, and CO₂ availability, resulting in a slow growth rate, and sub-optimal CO₂ uptake efficiency 124 (Iwasaki et al. 2021). In addition to the cultivation system, microalgal harvesting and product extraction 125 from harvested biomass are the challenges in the application of microalgae based carbon capture and 126 127 utilisations (Batan et al. 2013; Labeeuw et al. 2021b; Schädler et al. 2019).

This review aims to provide a comprehensive overview of the current state of microalgal culture developments. This review focuses on techniques to effectively deliver CO₂ and to enhance illumination of the microalga cells. The downstream process of culture (harvesting and water reuse) is also reviewed.
Finally, outcomes from a number of microalgae-based CCU projects are presented. This critical review will guide microalgal culture design to enhance biomass productivity and, thus, CO₂ capture. Advanced

development in microalgal culture design is expected to make microalgae-based CCU a viable option,

134 which can contribute to addressing the climate change challenge.

135 **2.** Carbon capture by microalgae

136 2.1 Current microalgal culture systems

The two most common systems for microalgal cultivation are open ponds (e.g. raceways) and closed photobioreactors (**Table 1**). Open ponds are the most simple microalgal cultivation systems. They have low capital and operating costs, and a low energy requirement. However, the open ponds are land intensive, and thus, are economically inefficient (Pugazhendhi et al. 2020). They are also sensitive to bacterial contaminations, high water evaporation rate, and challenging to maintain stable culture conditions.

143

[TABLE 1]

The closed photobioreactors have been developed to address the limitations of the open ponds. 144 145 The closed photobioreactors offer well-controlled culture conditions (e.g. temperature, pH, mixing, and contamination avoidance) and water loss prevention. The closed photobioreactors often have better 146 biomass productivity and space utilisation compared to the open systems. For example, tubular 147 photobioreactor achieved 2 to 2.5 folds higher biomass productivity compared to raceways under the 148 same conditions (Arbib et al. 2013). Areal biomass productivity of the floating film bag was at 21.1 149 150 $g/m^2/d$ compared to 8.1 and 5.9 $g/m^2/d$ in vertical tank reactors and raceways (Chinnasamy et al. 2010). 151 The closed photobioreactors can be constructed in a variety of configurations (Table 1). Transparent tubes, bags, or flat plates are used to make the closed photobioreactors. The tubes can be in vertical, 152 horizontal, and helical arrangements. Nevertheless, the closed photobioreactors presents a few 153 disadvantages. Bio-fouling (i.e. deposition of microalgae cells on tubes surfaces) is one major 154 155 drawback. The build-up biofilm prevents light penetration and illumination further into the middle of the culture tubes (Huang et al. 2017). Most of the available closed photobioreactors currently suffer 156 bio-fouling issues, requiring frequent system shutdown for cleaning (Katarzyna et al. 2015). Dissolved 157 oxygen build-up from the photosynthesis also inhibits biomass growth and thus reduces system 158

productivity (Kong et al. 2021; Singh & Sharma 2012). In this aspect, the closed photobioreactors often
require a degasser to reduce the dissolved oxygen below the inhibition threshold (Huang et al. 2017).
The closed photobioreactors also have high capital and operating costs (Huang et al. 2017). Currently,
the closed photobioreactors are mainly used for high-value compound productions (e.g. vitamins, amino
acids, and colorants).

There are several considerations when designing and selecting the types of microalgal culture systems (Singh & Sharma 2012; Tsoglin et al. 1996). Microalgal culture systems should provide optimal illumination (e.g. light illumination and availability) and high rates of CO₂ and dissolved oxygen transfer to prevent limitation in growth rates. The following section 3 and 4 will discuss recent developments to address the previously described disadvantages of microalgal culture systems.

169 2.2 Carbon fixation rate by microalgae

170 Carbon fixation rate by microalgae can be calculated based on the biomass production and171 biomass carbon content using the following equation (1):

172
$$R_{CO_2} = C_c \times P_B \times \frac{M_{CO_2}}{M_C} \qquad Eq \ (1)$$

173 Where: R_{CO_2} is the carbon fixation rate (mg/L.d); C_c is the carbon content (g/g of dry biomass); 174 M_{CO2} is the molecular weight of CO₂, Mc is the molecular weight of carbon; and P_B is the biomass 175 productivity (mg/L/d).

A significant number of studies has demonstrated carbon bio-fixation by microalgal cultures using air, CO₂-enriched air, or flue gases under both laboratory and industrial mass cultivation conditions (**Table 2**). The rate of carbon fixation varies significantly and depends on systems, operating conditions, and microalgal species used amongst studies.

180 CO₂-enriched air has been shown to enhance the productivity of microalgae under both laboratory
 181 and industrial mass culture conditions (**Table 3**). Duarte et al. (2020) achieved a 43 and 62% increase
 182 in specific growth rate and CO₂ bio-fixation efficiency when they supplied the tubular photobioreactor

183 with 10% v/v CO₂ gas at 0.05 L/L.min. Compared with ambient air, 1% v/v CO₂ gas achieved 60%
184 higher biomass productivity (Eloka-Eboka & Inambao 2017).

185 Current literature reviews also suggest the ability of microalgae to withstand high CO2 concentrations and combustion products (SO_x and NO_x) from flue gases (Table 3) (Vuppaladadiyam et 186 al. 2018). Yoo et al. (2010) compared the growth rate of Scenedesmus sp. and Botryococcus braunii 187 188 using flue gas and air-enriched CO₂. They achieved an increase in both biomass productivity and lipid content with flue gas. Likewise, Li et al. (2011) demonstrated the growth of Scenedesmus obliquus 189 190 using flue gas from a combustion chamber in a coke oven without treatment. These results significantly simplify CO₂ supply from flue gas. In some cases, the combustion products (SO_x and NO_x) can be 191 192 effectively used as nutrients for microalgae (Ho et al. 2017). Overall, direct application of flue gas could 193 potentially negate scrubbing systems at point sources (e.g. power plants).

194

- [TABLE 2]
- [TABLE 3]

196 3. Methods to introduce carbon dioxide to microalgae culture

197 Ideally, microalgal culture should not be limited by CO_2 availability to fully realize the potential of 198 microalgae for carbon capture. Current methods to introduce CO_2 into the culture system do not exceed 199 the CO_2 demands of the algae microalgae. Thus, the major challenge in the microalgal culture system 200 is to develop an efficient, scalable, and cost-effective carbonation system for high rate microalgal 201 growth to satisfy the scale for carbon capture requirement. This section will provide an overview of 202 different methods that have been used to date.

203 3.1. Sparging

Air, compressed CO₂, or flue gas is sparged into the microalgae culture via an air-diffuser at the bottom of raceways or closed photobioreactors. This basic method has low mass transfer efficiency due to the low retention time of gas bubbles in the culture medium (i.e. in a few seconds timeframe), pH, bubble size, and temperature (Duarte-Santos et al. 2016; Mendoza et al. 2013). Low CO₂ mass transfer also results in less CO₂ availability for microalgae fixation, loss of CO₂ back into the atmosphere, loss of energy, and associated cost with gas compressing and delivery.

Direct sparging gas into the culture via porous stones or air diffusers at the bottom of the raceway is 210 inefficient. Up to 90% of CO2 gas is not absorbed into the culture and is eventually vented back into the 211 atmosphere (Putt et al. 2011). Carbon supply can contribute 60% of operating costs at microalgae 212 213 cultivation plants. A direct injection may not be effective for CO₂ sequestration from flue gas (Langley et al. 2012). Micro and nano bubbles could enhance the dissolution of CO₂ into the culture both by 214 215 increasing the surface to volume ratio of the gas bubbles and the retention time (Temesgen et al. 2017). However, producing micro and nano bubbles is an energy-intensive process with high-pressure devices 216 (i.e. micro and nano bubble diffusers). In addition, the high shear stress induced by pressurized gas 217 218 bubbles could damage sensitive microalgal cells.

The introduction of a channel sump or a carbonation column is an alternative method to increase gas bubble retention time and, thus, CO₂ absorption (**Figure 1**). The channel sump can be external to or integrated into the raceways (i.e. airlift-driven raceway) (Fu et al. 2019; Ketheesan & Nirmalakhandan 222 2012). With the external channel sump, microalgae culture is delivered to mix with CO_2 gas at the 223 bottom of the channel sump. The greater the depth of the channel sump increases gas retention time (Fu 224 et al. 2019). In some instances, a mixer is introduced to increase the gas dissociation rate. Ketheesan et 225 al. (2012) utilized an airlift-driven raceway and achieved a CO_2 mass transfer of 33% when sparging 226 with a 0.25% CO_2 gas mixture. Due to the low mass transfer efficiency, purified CO_2 sources are not 227 recommended to be used with the sparging systems.

Carbonation column appears to be the most efficient sparging method to introduce CO₂. Microalgae culture is pumped into the carbonation column from the top to provide counter-current water flow to the gas bubbles (i.e. enter from the bottom) (**Figure 1**). The CO₂-rich microalgae culture is then transferred back to the raceway or the closed photobioreactors. Putt et al. (2011) appeared to be the first study using a carbonation column. In their study, microalgae culture was pumped at 7 L/min into a 3 m height column, and 5% CO₂ gas was supplied at 1.5 L/min. Putt et al. (2011) achieved 83% CO₂ transfer efficiency.

235

[FIGURE 1]

236

237 3.2. Bicarbonate solution

Dissolved inorganic carbon within culture medium consists of CO2, HCO3⁻ and CO3²⁻ with 238 concentrations depending on pH, alkalinity, salinity, and temperature (Iwasaki et al. 2021). All 239 microalgae can directly utilize CO_2 for photosynthesis. Many microalgae can also convert HCO_3^- to 240 CO_2 , while no microalgae are known to be able to use CO_3^{2-} . Thus, the pH of microalgae culture is often 241 242 maintained in the range of 7 to 8 to present dissolved inorganic carbon in the most usable form of CO₂ and HCO3⁻. The ability of microalgae to use HCO3⁻ opens a new way to provide a carbon source 243 (Abinandan & Shanthakumar 2016; Mokashi et al. 2016). Sodium bicarbonate has been introduced to 244 245 the Chaetoceros muelleri culture at levels from 0.25 to 1.0 g/L (Iwasaki et al. 2021). However, it is important to note that additional bicarbonate salts increase the alkalinity and salinity of the medium, 246 which may be toxic to microalgal culture. Previous studies have adapted alkaline tolerance species to 247

248 overcome this impact. Abiandan et al. (2016) demonstrated that C. pyrenoidosa could grow in 3.4 g/L sodium bicarbonate. Another potential approach is to combine bicarbonate solution and CO₂ gas for 249 carbon supply. First, bicarbonate solution increases the alkalinity of culture media. Under high 250 251 alkalinity levels, CO₂ gas dissociation can be enhanced. Qi et al. (2019) utilized CO₂ gas at 2% and 252 NaHCO₃ at 1 g/L to maintain culture pH at 7.7 for an optimal biomass and starch accumulation when 253 culturing Tetraselmis subcordiformis. It was observed that NaHCO3 addition alleviated the high dissolved CO₂ inhibition caused by the single CO₂ aeration and provided an effective carbon source 254 255 HCO₃⁻.

Although bicarbonate salt increases biomass production, its application in a full-scale culture system is limited due to its high cost compared to CO_2 gas. Recovery and reuse of the culture media after harvesting is possible to recover the carbonate. Nutrient levels must be balanced in these solutions for efficient algae assimilation, and nitrogen sources have different effects on pH stability. For example, nitrate has less of a pH effect than ammonium. To date, an industrial-scale application has not utilized this approach for carbon source supply in microalgal culture.

262 3.3. Carbonation via membranes

263 Membrane diffusers could increase gas-liquid interfacial area and contact time during CO₂ 264 supply. Membranes can be used in two different ways: a sparger or contactor devices (i.e. membrane sparger and membrane contactor). In the membrane sparging, CO_2 gas is pressurized via microporous 265 membranes to create a small gas bubble size (1-2 mm) compared to 5-8 mm with conventional sparging 266 267 systems to increase gas liquid mass transfer (Jana et al. 2017). Because of the small membrane pore size, high gas pressure is required in this system. Jane et al. (2017) applied gas pressure at 0.49 bar to 268 deliver CO₂ via a ceramic hydrophobic membrane. However, the current literature could not reaffirm 269 270 the benefits of membrane sparger over traditional sparging in terms of biomass production. Carvalho et 271 al. (2001) compared membrane sparger and traditional sparging in the cultivation of Nannochloropsis 272 sp. and showed a slight difference in biomass productivity. Moraes et al. (2020) utilized a hollow fiber membrane with a pore size of 0.4 μ m and membrane surface of 0.085 m² in a 1.7 L vertical tubular 273 274 photobioreactor for CO₂ sparing within the cultivation of *Spirulina* sp. LEB 18. The membrane sparger system provided a biomass concentration of 1.98 g/L, compared to 1.8 g/L in the conventional sparger after 15 days of culture (Moraes et al. 2020). It is also worth mentioning that the membranes require a large surface area and controlled gas pressure for CO_2 sparging. Membrane applications also are subjected to fouling (i.e. microalgal biofilm development on membrane surface), which requires frequent cleaning.

A membrane contactor or liquid-liquid membrane contactor is another membrane application for 280 delivery of CO₂ to microalgal cultures. In this approach, CO₂ is captured in a chemical solvent and 281 delivered to microalgal culture via a semipermeable membrane. Xu et al. (2019) combined a potassium 282 glycinate solution and hollow-fiber membrane for CO₂ delivery. CO₂ gas was loaded into the potassium 283 glycinate solution. Once this solution was saturated with CO2, it was circulated through the 284 285 semipermeable membrane to allow CO₂ transfer. The pH of the microalgal culture was used to regulate the CO₂ - rich solution. This configuration reduced the CO₂ loss (i.e. CO₂ utilisation efficiency of 90%) 286 while providing comparable biomass production with the conventional sparging systems (Xu et al. 287 2019). 288

289 **4.** M

4. Methods to enhance light exposure

An important consideration to optimise light illumination is the surface to volume ratio value (SA:V) of the culture system. In traditional reactor configurations (e.g. open ponds, raceway ponds, and closed photobioreactors), light utilisation is only optimal in a thin layer near the surface. More than 5 -10 cm away from the surface, illumination is negligible due to light attenuation and self-shading (Zavafer et al. 2021). At the surface, illumination is excessive and can inhibit photosynthesis (Iwasaki et al. 2021).

The closed photobioreactors have higher SA:V than the open systems. The SA:V of raceways or open ponds is in the range of 10-20 m⁻¹ depending on the depth of cultures (i.e. 5-20 cm). The closed photobioreactors have SA: V in the range of 25 to 125 m⁻¹ across different configurations (Eloka-Eboka & Inambao 2017). Amongst the closed photobioreactors, the helical type has SA:V of 53 m⁻¹ (Tredici & Zittelli 1998). Posten et al. (2009) changed the tube diameter and length to achieve different SA:V. Higher SA:V ratio of the closed photobioreactor also increases the illumination surface over a unit of footprint. However, increasing SA:V ratio increases the space required. Open ponds with low SA: V
would need significant land area for large-scale microalgal production and carbon capture. There are a
few approaches in practice or research to achieve an optimal light supply of all microalgal cells.

305 4.1. Thin-layer cascade

306 One notable characteristic of the thin-layer cascade (TLC) system is its high ratio of exposed surface to total culture volume (> 100 m⁻¹) (Celis-Plá et al. 2021; Grivalský et al. 2019). In the TLC 307 system, microalgae culture is distributed evenly at less than 5 cm at a flow rate of 0.4 to 0.5 m/s. This 308 configuration harnesses the benefits of open systems (i.e. direct light irradiance, easy heat diffusion, 309 310 rapid light/dark cycle, simple cleaning, and efficient degassing) as well as those of the closed 311 photobioreactors (i.e. high biomass densities and high volumetric productivity) (Grivalský et al. 2019). 312 The high biomass density contributes to the savings in the harvesting process. TLC system has 313 demonstrated high productivity with CO₂ gas supply. Marchin et al. (2015) achieved productivity of 24 and 10 g/m² d of dry biomass with and without 5% CO₂ gas dosing. These are equivalent to 48 and 20 314 315 $g/m^2 d$ of captured CO₂. However, the TLC system is not appropriate for large-scale application because 316 of its space requirements, sedimentation of microalgal cells, and energy-intensive and the high level of 317 CO₂ off-gassing in sparging method (de Marchin et al. 2015). The TLC system is not optimised for CO₂ dosing; CO₂ is dosed following the sparging method via an air stone (Section 3.1). 318

319 4.2. Submerged illumination

320 An internal illumination system was first developed in 1996 by Ogbonna et al. (1996). In this 321 system, a light source was inserted in a centrally fixed glass tube. More recently, internal illumination is typically designed for indoor cultivation. A notable feature of this system is that the lighting source 322 is placed vertically submerged into the culture solution. The light source is placed in a transparent tube 323 324 (e.g. opal acrylic light diffusing tubes and optical fibres) (Glemser et al. 2016; Xue et al. 2013). The 325 number of lights, light intensity, and positions can be selected to effectively utilise light with different microalgae species and culture conditions. Recently, Murray et al. (2017) has proposed an update on 326 327 the internal illumination where they developed the wirelessly powered, suspended, and free-moving LED. Their system contains a primary coil and seven secondary coil units with ferrite cores (i.e. wireless 328

329 light emitters) are enclosed in 30 mm diameter polystyrene housing. The primary coil was 22 cm in height, 15 cm in width, and contained 290 wire turnings. Biomass yield per mol of photons was 330 significantly higher in their system than the external illumination at the same light intensity. The yield 331 on photons was 1.18 vs 0.78 and 1.15 vs 0.05 g biomass per mol photons for C. vulgaris and H. pluvialis, 332 333 respectively, in submerged-light photobioreactor vs external illumination (Murray et al. 2017). The submerged-light photobioreactor provided more uniform light to the culture and reduced dark zones 334 and the self-shading effect. Recently, side-emitting optical fiber has also been tried for enhancing the 335 336 microalgae biomass productivity. Wondraczek et al. (2019) enhanced the illumination efficiency by 337 more than two orders by applying the side-emitting optical fiber within culture. The production of green 338 algae Haematococcus pluvialis was increased by 93%. Side-emitting fiber also allows modifying the 339 lighting spectrum into different reactor configurations. However, the authors have also identified the 340 number of challenges to upscale this technology in microalgae-based carbon capture. Biofouling on the lights is significant and impairs the light distribution, especially at high biomass levels. Intensive mixing 341 is also required to keep the wireless light emitters in suspension. It is also a challenge to maintain the 342 343 wireless light emitters at a designed location in the culture system. Nevertheless, this promising 344 technique is still in its infancy. Advanced development in biofilm resistant material could reduce 345 biofouling (Demetz et al. 2020; Murray et al. 2017). Adding more wireless light emitters would also reduce intensive mixing when balancing the costs of light and mixing systems. The submerged 346 illumination is only limited to indoor closed cultivation using artificial light. Coupling with advanced 347 development in PV power to the system will have benefits in reduction of carbon footprint. 348

349

350 4.3. Airlift-loop reactor

The airlift-loop reactor is a patent by Subitec cultivation technology. Its design is based on the principle of an airlift-loop reactor to achieve an optimal light supply to all microalgal cells due to the thin layer thickness and complete intermixing and homogenization by static mixer. This design avoids many of the previously described disadvantages (e.g. photo inhibition, shear stress on microalga cells) of the traditional closed photobioreactor.

356 5. Downstream processing

The pathways to utilise microalgal biomass will determine the contribution of microalgae-based 357 358 CCU on either net-zero or negative emissions. Microalgae biomass can be used to replace nonrenewable fossil resources to produce chemical precursors, biopolymers, personal care products, 359 360 nutraceuticals, agricultural feed, and fuels. In this pathway, the captured CO_2 will be released back to 361 the atmosphere and continue to be captured in the next cycle. This results in net-zero emissions. Another 362 pathway is converting microalgal biomass into a long-term product for carbon storage. Bioplastic production and mixed in cement are potential applications of microalgal biomass for long-term storage 363 364 (Section 5.2.2). Apart from biomass utilisation, effective microalgae harvesting methods and water reuse will reduce operating costs and enhance the carbon capture in the microalgae-based CCU 365 366 technology.

367 5.1. Microalgae harvesting

Cost-effective microalgal biomass harvesting along with a profitable revenue stream (utilisation) is probably required to offset the capital and operating cost and make microalgae-based carbon capture financially competitive. Microalgae are tiny (i.e. less than 100 μm in size) with a negative charge surface, grow in dilute and suspended culture media, and have a similar density to water. These properties cause challenges in liquid-solid separation techniques to harvest microalgae effectively (Labeeuw et al. 2021b; Nguyen et al. 2019).

374 Microalgae harvesting techniques that are utilized at an industry scale includes centrifugation, membrane filtration, flocculation, sedimentation, and flotation (Barros et al. 2015; Fasaei et al. 2018). 375 376 Centrifugation is a mechanical technique, which exerts a centrifugal force on the algal cells to separate 377 them from the growth medium. Despite being a highly effective technique with >90% efficiency, 378 centrifugation requires expensive investment costs for equipment and high-energy consumption (Dassey & Theegala 2013; Najjar & Abu-Shamleh 2020). Membrane filtration is an emerging 379 harvesting technology that preserves cell quality and does not require chemical addition. However, the 380 accumulation of microalgae and extracellular organic matter on the membrane over time causes 381 membrane fouling, thus necessitating a high cost of membrane cleaning and replacement (Barros et al. 382

383 2015). Flocculation has been proposed as a low-cost and effective harvesting technique via three mechanisms: charge neutralization, bridging, and sweeping effect. This technique offers a short and 384 385 simple operation procedure with minimal energy demand, thus being suitable to harvest a wide range of microalgae at large-scale production (Barros et al. 2015). Sedimentation requires a longer retention 386 387 time to harvest the microalgae and has lower efficiency than other techniques (Figure 2). The harvesting efficiency can be increased by combining sedimentation with flocculation. Flotation utilizes air or gas 388 389 bubbles to carry the tiny microalgal cells to the water surface. The main advantages of flotation are 390 short operation time, compactness, large-scale harvesting, and high flexibility with low initial cost 391 (Laamanen et al. 2016; Singh & Patidar 2018). Research and development in synthetic flocculants (e.g. 392 polymers) could produce new polymers with tuneable properties for microalgae harvesting to minimize 393 the harvesting cost (Li et al. 2017; Liu et al. 2018).

The selection of harvesting methods depends on a number of criteria, including techno-economic 394 efficiency, type of microalga species, and intended biomass application (Figure 2). The techno-395 economic efficiency of a harvesting technique is evaluated based on factors such as processing time, 396 397 capital and operation cost, scalability and integration with the existing culture systems. For example, 398 flocculation can achieve >95% harvesting efficiency with simple equipment and low energy usage (Vu et al. 2021). This is likely to be more techno-economically efficient for large-scale operation than using 399 400 membrane filtration, which requires high capital and operational cost to maintain the membrane (Fasaei 401 et al. 2018). The selection of harvesting techniques is also species-specific, as this can influence the 402 quality of harvested biomass and subsequent use of biomass. Species with a rigid cell wall (e.g. 403 Chlorella and Nannochloropsis oculata) are resistant to shock and damage caused by harvesting 404 techniques (Sales et al. 2019). On the other hand, species without a rigid cell wall (e.g. Porphyridium) 405 are prone to cell damage and cell rupture (Labeeuw et al. 2021b). High-value applications (e.g. 406 biochemical and biopolymers) require the microalgae to retain their cellular structure post-harvesting, 407 which is possible via centrifugation and membrane filtration. Flocculation can cause contamination of the biomass due to the presence of chemical flocculants, which may not be suitable for high-value 408 409 applications. However, as this issue is insignificant in biofuel application, flocculation with its superior

410 techno-economic efficiency is the most suitable harvesting technique for biofuel production from411 microalgae.

412

[FIGURE 2]

413

414 5.2 Utilisation of biomass for carbon storage

415 Biomass utilisation in the microalgae-based carbon capture process refers to the ability to convert 416 microalgal biomass into value-added products that otherwise need to be extracted from fossil sources. Through photosynthesis, microalgae convert inorganic carbon into macromolecules, metabolites, and 417 biochemical compounds (e.g. lipids, carbohydrates, and proteins). Microalgae-based carbon capture and 418 utilisation could simultaneously mitigate climate change and generate commercial products. Microalgae 419 420 also can offset or reduce carbon by replacing items currently derived from carbon-intensive practices using fossil fuel. The concept of carbon capture and utilisation via a microalgal biorefinery is in line 421 with the IEA's Task 42 objective "Biorefining in a circular economy" and CO₂ utilisation in a circular 422 423 economy (Srinivasan et al. 2021).

424 5.2.1 Displacing non-renewable fossil resources

There are many products that can be made with the whole algal biomass without further processing (i.e. after harvesting and drying stages). These include food (e.g. *Spirulina* supplements) (Sutherland et al. 2021), energy (e.g. syngas or bio-oil), soil additives (e.g. biostimulants), or feedstock for anaerobic digestion (Choi et al. 2019).

Human food is limited to a select number of species. Still, it can have several nutritional benefits in the diet, such as an improved source of protein, vitamins, and various omega fatty acids (Wells et al. 2017). Functional food from algae can have lower environmental impacts from producing the exact nutritional amounts via traditional species (Ye et al. 2018). There is a wide range of algal species that could be used as a feedstock supplement, both for aquaculture and ruminant feedstock. Increased microalgal production could support farmed aquaculture by replacing the current energy-intensive feed products used (e.g. animal by-products), or increasing the efficiency of farmed fish (i.e. reducing the 436 stressors on the ecosystem and fuel use from wild-catch) (Henriksson et al. 2013), or could be an alternative source of the polyunsaturated fatty acids (PUFA), removing of fish altogether as a source of 437 438 oil (Beal et al. 2018). One study has investigated the potential of algae combined with a CCS system to provide both fishmeal and carbon storage (Beal et al. 2018). Algae have also been shown to promise 439 440 ruminant feedstocks, both in having positive effects on the diet and quality of the product (Altomonte 441 et al. 2018; Madeira et al. 2017). This could potentially lead to lower demand for current agricultural 442 feedstocks (Amorim et al. 2021). Algae could also significantly reduce the methane output of the 443 ruminants by altering the diet (at low levels of inclusion), leading to lower levels of carbon in the 444 atmosphere (McCauley et al. 2020).

Algae have a lot of potential as an energy source. This could be either through the extraction of the 445 446 high lipid content of some species for biodiesel or bioethanol or through thermal treatment (e.g. gasification, or pyrolysis) or anaerobic digestion of the whole cell to produce bio- or syngas (Choi et 447 al. 2019). These have the advantages of replacing fossil fuel alternatives but are currently not cost-448 competitive at scale and require further research to reduce costs (t Lam et al. 2018). Anaerobic digestion 449 450 (AD) of the whole cell, however, has several advantages – it can produce energy (biogas) while also 451 producing several products of interest (such as organic acids) (Kassim & Meng 2017). AD also has the 452 advantage that it can be used on the residual biomass leftover from various extraction processes.

Moreover, various products of interest can be produced from microalgal components. These can include beta-carotene and other carotenoids, pigments, proteins, the extracted PUFA, among others. These can be already be found in various cosmetics and nutraceuticals (Borowitzka 2013). Often these compounds make up only a tiny portion of the overall cell (e.g. 1-10%), so a biorefinery approach extracting multiple products of interest increases the overall economic feasibility of the system (t Lam et al. 2018).

459 5.2.2 Permanent carbon storage

460 Microalgae can potentially be used in several long-term products, such as mixed in cement or
461 made in bioplastics (Li et al. 2016), which may be qualified as carbon sequestration/storage techniques.
462 Another potential usage of microalgae is the use of algal biochar, which can be added to the soil for

463 more long term carbon storage (Sayre 2010) and moving towards sustainable agriculture (Sutherland et
464 al. 2021). Algae could also be used as biostimulants to improve crop production, thereby reducing the
465 need for fossil-based fertilisers (Mona et al. 2021).

466 Plastics are currently produced from fossil sources and are difficult to break down (Zhang et al. 467 2019). Bioplastics can either come from a biological origin, or be biodegradable, or both. Microalgae can have a high proportion of polysaccharides, proteins, or lipids, which all can feed into the current 468 469 bioplastic processing systems and produce either more traditional plastics or biodegradable plastics 470 such as polylactic acid and polyhydroxyalkanoates (Zhang et al. 2019). Producing plastic from 471 biological origin could reduce GHG emissions by 67 - 116% compared to traditional sources (Beckstrom et al. 2020). Bioplastics that can biodegrade (back to CO_2 and water) can also reduce the 472 473 impact on the ocean and landfills that are currently being stressed by non-degradable plastics. However, some plastics produced from microalgae that are more durable could act as carbon sinks. 474

Another possible long-term carbon sink is the use of microalgae in cement. This relies on carbonate precipitation (CaCO₃) that can be performed by some photosynthetic microalgae or cyanobacteria, as well as non-phototrophic bacteria (Alshalif et al. 2020). Cyanobacteria are of particular interest in this process as they have been shown to be able to precipitate the carbonate intracellular (Castro-Alonso et al. 2019). There has been some evidence suggesting that the overall durability of the product could be improved by incorporating microalgae for carbonate precipitation.

481 5.3. Water reuse

482 Microalgal biomass production facilities have large water footprints. For example, a closed tubular photobioreactor has a total water footprint in the range of 2.4 to 6.8 m³/kg of dry biomass (Batan 483 et al. 2013; Farooq et al. 2015; Martins et al. 2018). Batan et al. (2013) conducted a life cycle assessment 484 and reported the water footprint of the microalgae-based biofuel facility within $23 - 85 \text{ m}^3/\text{GJ}$. Yang et 485 al. (2011) reported a value of 3 m³/kg of biodiesel. The high water footprint is a roadblock for large-486 scale microalgal production in places where sunlight is abundant (e.g. deserts) (Venteris et al. 2013). In 487 recognition of the water footprint, there is a rising interest in culture media recycling. Recycling of 488 culture media reduces water consumption and the environmental impact of microalga cultivation. 489

490 Recycling the culture residual can save up to 80% of water requirements and reduce nutrient input (Fret
491 et al. 2017; Yang et al. 2011). Yang et al. (2011) obtained a 55% reduction in nutrient requirement when
492 recycling culture media after harvesting.

493 Before reuse, the culture media after harvesting may contain many growth inhibitors that needed to be pre-treated (Zhang et al. 2016). The types of growth inhibitors present in the culture media are 494 subject to harvesting methods (e.g., centrifuge, flocculation, and filtration) and microalgal species 495 (Labeeuw et al. 2021a). Typical inhibitors include cell wall debris, bacteria, algal organic matter 496 (AOM), coagulants, flocculants, and accumulated micronutrients (Fret et al. 2020; Ganuza et al. 2016; 497 498 Monte et al. 2019; Zhang et al. 2016). Cell wall and other cell debris derived from cell division can cause aggregation of both algae and bacteria. Bacteria that may grow well in recycled media (if not 499 sterilized) can compete with microalgae for nutrients. The presence of AOM in the residual culture can 500 501 facilitate the growth of bacteria, thus affecting the subsequent microalgae growth. Moreover, AOM, 502 especially allogenic organic matter (e.g. polysaccharides, humic and fulvic-like substances, and crude ethyl acetate) found in the culture media after harvesting can reduce the algal growth rate and 503 photosynthetic efficiency (Sha et al. 2019). Residual alum or ferric ions, components of flocculants, are 504 likely toxic to microalgae in the recycled media. Unbalanced consumption of different ions (i.e. NO₃, 505 PO_4^{3-} , Na^+ , K^+ , Ca^{2+} , and Mg^{2+}) for the microalgae growth causes elevated salinity in the medium 506 solution after harvesting. Salinity of nutrient salts significantly affects biomass productivity via osmotic 507 508 stress. Thus, a pre-treatment step is often required before replenishment to the culture system to 509 minimize the aforementioned negative impacts of growth inhibitors. Current pre-treatment technologies 510 can be classified based on two major target inhibitors (i.e. bacteria and inhibitory dissolved organic matters). 511

The culture residual is the result of microalgae harvesting. In most cases, its quality depends on the harvesting methods (Labeeuw et al. 2021b). For example, flocculation with inorganic salts (e.g. FeCl₃ and AlCl₃) produced high residual levels of Fe^{3+} and Al^{3+} in the culture residual. In addition, the culture residual contains leftover nutrients, cells debris, bacteria, and algal organic matters. Thus, it is often 516 required a treatment step before replenishment to the culture system. Current treatment methods are 517 mainly UV sterilisation, and filtration.

518 5.3.1 UV/Ozone and adsorption

Algal organic matter (AOM) in the residual culture can be degraded into harmless fractions (e.g. nutrients) using advanced oxidation to minimize its inhibitory effects on the microalgae growth. Hydroxyl radicals released from an advanced oxidation process can effectively oxidize and mineralize organic matter, thus producing more biodegradable substances. UV-based advanced oxidation processes (i.e. UV/H₂O₂, UV/peroxydisulfate, and UV/NH₂Cl) could effectively degrade AOM and convert the growth inhibitors in the residual culture into a nutrient source for the growth of *Scenedesmus acuminatus GT-2* and *Dunaliella salina* (Wang et al. 2018).

Extraction of AOM from the residual culture using adsorption and flocculation has been extensively applied in several laboratory-scale studies (Mejia-da-Silva et al. 2018; Morocho-Jácome et al. 2015; Morocho-Jácome et al. 2016; Sha et al. 2019; Zhang et al. 2016). Granular activated carbon (GAC) and powdered activated carbon (PAC) are two popular absorbents used to remove AOM from the residual culture (Morocho-Jácome et al. 2016; Sha et al. 2019).

531 5.3.2 Membrane filtration

532 Membrane filtration has emerged as the most effective technique to sterilize the residual culture (Pugazhendhi et al. 2020). Membrane filtration can be either integrated with a harvesting step (Fret et 533 al. 2017; Monte et al. 2019; Sha et al. 2019) or used as an independent process for removing bacteria 534 and particulate matter from the residual culture after harvesting (Farooq et al. 2015; Fret et al. 2017; 535 536 Fret et al. 2020). Fret et al. [35] demonstrated that after pre-treatment by microfiltration, the residual 537 culture could be used to cultivate successfully Nannochloropsis sp. and Tisochrysis lutea without crosscontamination. The authors could save up to 80% of water consumption during 167 days of the 538 539 cultivation.

540 In addition to the membrane filtration, sand filtration and other disinfection methods (i.e. sand 541 filtration, ozonation, UV, chlorination, and heating) have proved their efficacy in eliminating non542 beneficial bacteria in the recycled media (Fret et al. 2020; Ganuza et al. 2016; González-López et al. 2013; Monte et al. 2019). Sand filtration is utilized as either a pre-treatment step before the membrane 543 filtration (i.e. microfiltration and ultrafiltration) (Fret et al. 2017) or a post-treatment step after 544 flocculation and coagulation processes (Fret et al. 2016). A study conducted by González-López et al. 545 546 [39] showed that ozonation was most effective in removing bacteria from the residual culture to grow 547 Nannochloropsis gaditana. Another novel robust method that uses pH-induced flocculation followed 548 by sand filtration to remove non-beneficial bacteria from the residual culture for C. vulgaris growth was 549 developed and demonstrated by Fret et al. [34]. To date, culture media recycling has been applied mostly 550 in lab-scale and a few pilot-scale cultivations (Fret et al. 2017; Fret et al. 2020).

551 5.4. Cost of microalgae-based carbon capture and utilisation

552 The cost of microalgae-based CCU depends on several parameters (e.g. productivity of culture systems and utilisation pathway). Results from some demonstration projects have indicated the high 553 554 cost of microalgae-based carbon capture. The joint project between the Center for Applied Energy 555 Research and Duke Energy used a closed photobioreactor design to test the CO₂ capture from the East Bend Station. The project considered CO₂ capture for coal-fired power plants, production of biofuels, 556 and other bioproducts from microalgal biomass. The project used flue gas after the scrubber and the 557 selective catalytic reduction treatments. The vertical tube photobioreactors provided 18 m³ culture 558 volume with a 19 m³ feed tank and 5.7 m³ harvest tank. The average daily productivity was slightly 559 above 10 g/m²/d. The project provided an estimated US\$ 1451.5 per tonne of CO₂ over 10 years period 560 (Wilson et al. 2014). The primary cost contributed from culture system and installation. In comparison, 561 direct air carbon capture using a conventional liquid-based absorption process is at the cost of US\$ 162 562 563 to 387 per tonne of CO₂ (Srinivasan et al. 2021).

Microalgae-based carbon capture and utilisation also require a large amount of land. In a demonstration project at Bayswater coal-fired power station in Australia, Burgess et al. (Burgess. et al. 2011) concluded that solar-powered photobioreactor could capture 25-50% of the CO₂ emitted from the power station. A maximum capture rate was around 30 g/m²/d, which required 150 Km² of bioreactor surface area to treat the flue gas output from one of the 4 x 600 MW units (Burgess. et al. 2011). 569 Seambiotic conducted a demonstration project at the Rutenberg Power Station in Israel. The project used open ponds with a total surface area of 1000 m² and flue gas with 12% CO₂ content. The 570 harvested biomass was a dry algae powder, which was used in fish and animal feed supplements. They 571 achieved an average biomass production of $20 \text{ g/m}^2/\text{d}$ (Seambiotic Ltd, 2010). To date, Seambiotic Ltd 572 573 provides significant quantities of microalgal biomass using flue gas from a coal-fired power plant. Although the cost of carbon capture is not available, the utilisation of harvested biomass for high-value 574 products could provide positive revenue. In comparison to other CCS, microalgae-based carbon capture 575 576 and utilisation should be viewed as a pathway to producing high-value products.

577 6. New perspectives and directions

578 Microalgae-based CCU is a promising alternative to conventional CCU. The captured CO₂ in the 579 form of microalgal biomass could be utilized to produce valuable products. In comparison to chemical 580 and physical CO₂ capture processes, the microalgae-based capture is environmentally sustainable. 581 However, microalgae-based carbon capture and utilisation is a complex process. Many inter-connected 582 parameters such as species, culture conditions, CO₂ concentration, pH, temperature, irradiance, and 583 culture systems influence carbon fixation rate. Amongst these, new cultivation systems should optimize 584 the distribution of light, nutrients, and carbon dioxide for large-scale carbon capture.

585 Microalgae-based CCU should not compete with agriculture for nutrients (i.e. fertilizer). 586 Sourcing nutrients from a waste stream such as wastewater treatment plants (e.g. secondary effluent or 587 anaerobic digestate) for microalgal culture would reduce the operating cost, while also providing a 588 pathway to treat wastewater. The feasibility of using wastewater as nutrients source has been well tested 589 in the literature. However, the integration of wastewater into microalgae-based carbon capture and 590 utilisation has not been explored. It is anticipated that the applied method in culture media recycling 591 (Section 5.3) could be used to prepare the wastewater.

592 Microalgae harvesting is a crucial step in microalgae-based CCU. The selection of harvesting 593 method determines operating costs and the biomass utilisation pathways. Amongst the current 594 harvesting methods, flocculation has emerged as a versatile microalgae harvesting method considering key selection criteria such as scalability, biomass quality, operating cost, processing time, and intended biomass applications. The performance of flocculation strongly depends on the types of flocculants and microalgae species. Microalgae cells are usually suspended particles with a negative surface charge. Thus, polymers for microalgae harvesting are usually cationic (i.e. positively charged) to neutralize the negative charge of the microalga cells. New methods in polymer synthesis (e.g. free-radical polymerisation) could be used to produce cost-effective flocculants specifically for microalgae-based CCU.

602 The economics of microalgae-based CCU can be significantly improved if the harvested products 603 can be commercialized. Therefore, high-value strains to produce commercially useful applications are also a key to promoting microalgae capture of CO_2 . The selections should focus on the fast growth rate, 604 high photosynthetic rate, and strong environmental tolerance species with simple downstream 605 processing (e.g. harvesting and water reuse). The selected species should potentially produce high-value 606 607 biomass. Currently, microalgae species, which can grow well with flue gas supply, do not often have high commercial value. However, microalgae are a highly diverse group of microorganisms (c.a 0.2 -608 1 million recognized species), which provide a significant biobank for selection. In addition, the 609 development of microalgae biotechnology (e.g. genetic modifying organisms) can be applied to enhance 610 611 CO₂ fixation in future research.

612 7. Conclusion

613 Microalgae-based CCU is technically feasible using the current open ponds or the closed 614 photobioreactors. However, the economic feasibility of microalgae-based CCU is still a significant 615 challenge. This is partially due to the limitations of microalgal culture. They are not specifically 616 designed for high throughput carbon capture and thus, have limitations (e.g., not optimal lighting 617 distribution, nutrient, and CO₂ supply). Open ponds have poor space utilisation and are not viable for 618 large-scale carbon capture. The closed photobioreactors have high capital and operation costs. 619 Techniques to intensify microalgae cultivation for large-scale carbon capture are needed. The 620 downstream processes of microalgae culture (i.e. harvesting, biomass utilisation, and water reuse) are

- 621 crucial to the economic feasibility. Collectively, microalgae-based CCU can be viewed as a pathway to
- 622 generate high-value products until a high-rate culture system becomes available.

623 E-SUPPLEMENTARY FILES OF THIS WORK CAN BE FOUND IN ONLINE VERSION OF

624 THE PAPER.

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1012 List of table

1013 Table 1: Summary of different open and closed cultivation systems with their advantages and

1014 disadvantages.

Systems	Advantages	Disadvantages	Ref
Open system	15		
Circular pond	 Low capital cost Simple operation Easy construction Dissolved oxygen discharge 	 Low efficiency Uncontrollable cultural conditions High evaporation rate High risk of contamination Poor space utilisation Uneconomical for scale-up Low surface to volume ratio (SA:V) 	(Eloka-Eboka & Inambao 2017; Kong et al. 2021; Vo et al. 2019)
Raceway pond or high rate algal pond	 Low capital cost Simple operation Easy construction Dissolved oxygen discharge 	 Low efficiency Uncontrollable cultural conditions High evaporation rate High risk of contamination Poor space utilisation Uneconomical for scale-up Low surface to volume ratio (SA:V) 	(Eloka-Eboka & Inambao 2017; Kong et al. 2021; Vo et al. 2019)
Closed phot	obioreactors		
Horizontal tube PRB	 Stable performance High productivity Avoid contamination Easy to maintain operating conditions Less hydrodynamic stress 	 High capital cost Dissolved oxygen accumulation High energy for pumping Bio-fouling on surface High surface to volume ratio 	(Eloka-Eboka & Inambao 2017; Tsoglin et al. 1996)
Vertical tube PRB (bubble and airlift types)	 Improved gas/liquid transfer High CO₂ dissociation rate Dissolved oxygen discharge 	 High capital and operating costs Challenge to clean up Low economic viability 	(Arbib et al. 2013; Kong et al. 2021)
Flat plate PBR	 High illumination surface overland size High SA:V ratio Avoid contamination Dissolved oxygen discharge Less energy consumption 	 High capital cost Large scale systems need significant support materials Difficult to regulate temperature Difficult to maintain cells in suspension Bio-fouling formation 	(Ho et al. 2017)
Floating film bag PBR	 Low cost for construction Avoid contamination Simple design 	 Poor mixing Difficult to maintain cells in suspension Difficult to clean up Leakage Bio-fouling formation Short life span 	(Chinnasamy et al. 2010; Labeeuw et al. 2021b)
Helical type PBR	- Optimize SA:V ratio	- High capital and operating costs	(Singh & Sharma 2012;

- High illumination	- High energy consumption for	Tsoglin et al.
surface	mixing culture	1996)
- Optimize land use	- High hydrodynamic stress on cells	
- Avoid contamination	- Bio-fouling formation	
- High CO ₂ dissociation	e	
rate		
- Dissolved oxygen		
discharge		

Species	CO ₂ supply (%, v/v)	Supply method	Scale	CO ₂ removal efficiency (%)	CO ₂ fixation rate (g/L.d)	Biomass produced (g/L. d)	References
Botryococcus braunii	0.03	Sparging	1 L glass bottle	-	0.08*	0.04	(Rodas- Zuluaga et al. 2021)
Botryococcus braunii	10	Sparging	1 L glass bottle	6.78 ± 3.58	0.03*	0.02	(Rodas- Zuluaga et al. 2021)
Botryococcus braunii	20	Sparging	1 L glass bottle	3.73 ± 0.74	0.05*	0.03	(Rodas- Zuluaga et al. 2021)
Scenedesmus sp.	20	Sparging	1 L glass bottle	3.82 ± 1.71	0.23*	0.13	(Rodas- Zuluaga et al. 2021)
Chlorella vulgaris	0.03	Sparging	1.5 L membrane bioreactor	-	0.09	0.05	(Nguyen et al. 2020)
Chlorella sorokiniana	1	Sparging	2 L flask	-	0.58	0.29	(Ding et al. 2020)
Chlorella pyrenoidosa	1	Sparging	2 L flask	-	0.49	0.24	(Ding et al. 2020)
Scenedesmus obliquus	10	Sparging	1.8 L bubble column	94.7	0.27*	0.15	(Liu et al. 2020)
Chlorella pyrenoidosa	10	Sparging	1.8 L bubble column	95.1	0.25*	0.14	(Liu et al. 2020)
Scenedesmus dimorphu	10	Sparging	1.8 L bubble column	94.6	0.22*	0.12	(Liu et al. 2020)
Chlorella vulgaris	10	Sparging	1.8 L bubble column	95.3	0.13*	0.07	(Liu et al. 2020)

1016	Table 2: Biomass	production by	various microa	lgae species at	different CO ₂ conten	ts and supply methods
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Spirulina platensis	2.5	Injection	250 L bubble columns in - series	0.19	0.12	(Almomani et al. 2019)
Chlorella fusca	0.03	Injection	1.7 L tubular column	0.21	0.15	(Deamici et al. 2019)
Chlorella vulgaris	5	Sparging	0.25 L column	0.74	0.39	(Al Ketife et al. 2017)

1017 *The CO₂ fixation rate is calculated based on the assumption that 1.8 kg of CO₂ is fixed to produce 1 kg of microalgae biomass.

CO ₂ source	Gas composition (%)	Microalgae	Photobioreactor	CO ₂ removal efficiency (%)	CO ₂ fixation rate (g/L d)	Biomass production (g/L.d)	Reference
Co-generation power plant	CO ₂ (3-6) O ₂ (12)	Haematococcus pluvialis	6 L tubular, vertical PBR	-	0.13*	0.07	(Choi et al. 2017)
Combustion of coal	$\begin{array}{c} CO_2 (10 \pm 2) \\ O_2 (8) \end{array}$	Chlorella vulgaris	Bubble column	-	0.34	0.37	(Yadav et al. 2021)
Combustion of coal	$\begin{array}{l} \text{CO}_2 \ (10 \pm 2) \\ \text{O}_2 \ (8) \ \text{NO}_2 \ (0.0061) \\ \text{SO}_x \ (0.003) \end{array}$	Chlorella vulgaris	0.5 L Bubble column	-	0.15	0.17	(Yadav et al. 2019)
Coal burning boiler outlet	$\begin{array}{c} CO_2 (7) \\ NO_x (0.021) \\ SO_x (0.012) \end{array}$	Scenedesmus quadricauda	0.5 L Flask	81	0.43*	0.24	(Kandimalla et al. 2016)
Coal burning boiler outlet	$\begin{array}{c} \text{CO}_2 (6) \\ \text{NO}_x (0.025) \\ \text{SO}_x (0.018) \end{array}$	Chlorella vulgaris	0.5 L Flask	72	0.14*	0.08	(Kumar et al. 2018)
Coke oven in steel pant	$\begin{array}{c} \text{CO}_2 (25) \\ \text{O}_2 (6 - 8) \text{ NO}_x (0.0075) \\ \text{SO}_2 (0.0085) \end{array}$	<i>Chlorella</i> sp.	1 L bubble column	16	0.88	0.52	(Kao et al. 2014)
Oil producing industry	$\begin{array}{c} \text{CO}_2 \ (15.6) \\ \text{CH}_4 \ (10.6) \\ \text{N}_2 \ (72.8) \\ \text{H}_2 \text{S} \ (0.012) \end{array}$	Chlorella sorokiniana	1.8 L serially connected airlift and bubble columns	4.1	0.41*	0.23	(Kumar et al. 2014)
Simulated flue gas	$\begin{array}{c} \text{CO}_2(15) \\ \text{N}_2 \ (85) \\ \text{NO}_x \ (0.001) \ \text{SO}_2 \ (0.002) \end{array}$	Desmodesmus armatus	0.25 L bubble column	-	2.34*	1.30	(Guo et al. 2017)

Table 3: Microalgae-based carbon capture from point sources (i.e. industrial flue gas)

1019 *The CO₂ fixation rate is calculated based on the assumption that 1.8 kg of CO₂ is fixed to produce 1 kg of microalgae biomass.

1020	List of Figures
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[FIGURE 1]

Figure 1: The schematic diagram of different bubbling or sparging to introduce CO₂ in microalgae

1023 culture (e.g. raceways) (1) direct bubbling, (2) a channel sump, (3) airlift-driven raceway (other names:

- 1024 internal loop airlift reactor), and (4) carbonation column.
- 1025

[FIGURE 2]

- 1026
- 1027 Figure 2: Microalgae harvesting method selection criteria (a) and relative comparison of different1028 methods (b).